CRD

# Nanoscience on High Performance Computers: From Methods to Codes to Applications

(Plane-wave, DFT codes)


**Andrew Canning**

**Computational Research Division**

**LBNL**



(Funded by DOE computational nanoscience initiative (MICS, BES))


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# Outline



- Introduction to Nanoscience
- Electronic Structure Calculations (DFT)
- Code performance on High Performance Computers
- New Methods and Applications

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# DOE initiative:Theory Modeling and Simulation in Nanaoscinece

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## Scalable Methods for Electronic Excitations and Optical Responses of Nanostructures: Mathematics to Algorithms to Observables

Lead PIs: Juan Meza (LBNL) Martin Head-Gordon (UCB,LBNL)
(Steven Louie, Lin-Wang Wang, Andrew Canning, John Bell, Chao Yang, Chuck Rendleman at LBNL, Emily Carter(Princeton), James Chelikowsky (UMN) John Dennis (Rice University), Yousef Saad (UMN) )

## Predicting the Electronic Properties of 3D, Million-Atom Semiconductor Nanostructure Architectures

Lead PIs: Alex Zunger (NREL), Jack Dongarra (UT, ORNL)
(Lin-Wang Wang, Andrew Canning, Osni Marques at LBNL, A. Franceschetti, W. Jones, K. Kim, G. Bester NREL)

Projects funded jointly by MICS and BES in DOE

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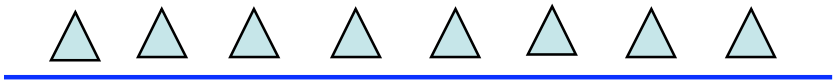
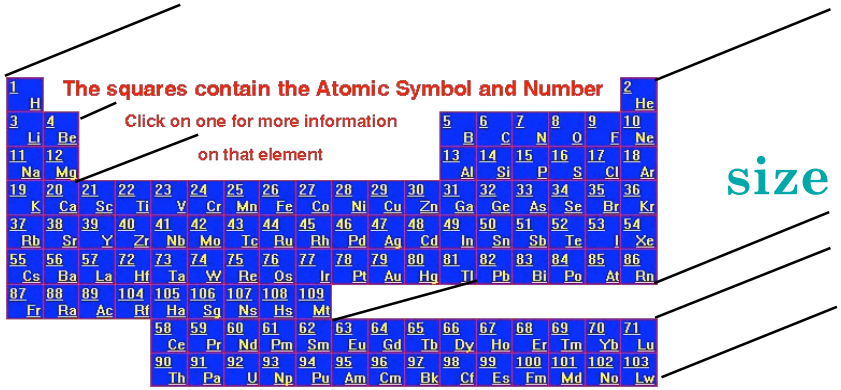
# Nanostructures as a new material

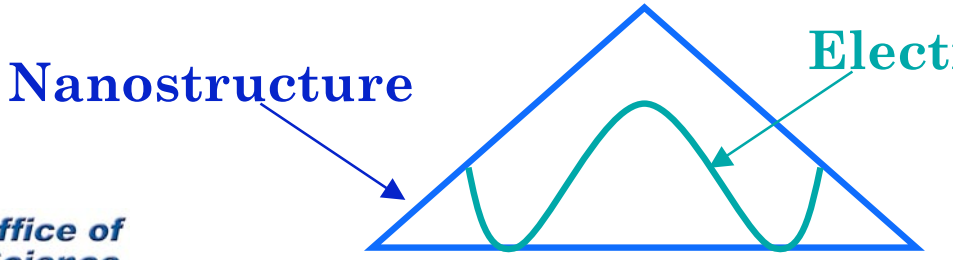


**Definition:** **Nanostructure is an assembly of nanometer scale "building blocks".**



**Why nanometer scale:** **This is the scale where the properties of these "building blocks" become different from bulk.**



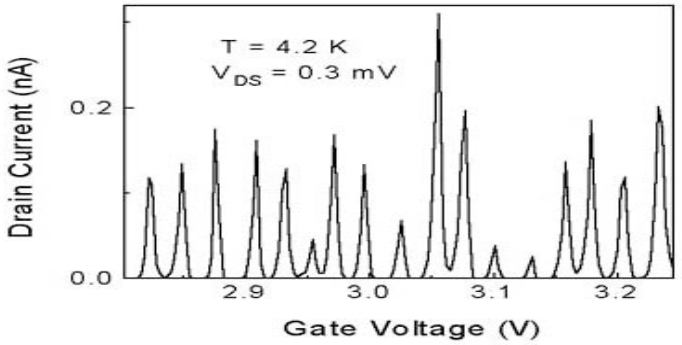

**Both are in nanometers**

# CRD Example: Quantum Dots (QD) CdSe

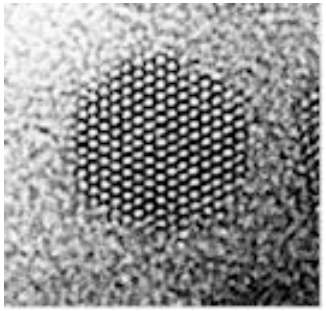
- Band gap increase


CdSe quantum dot (size)

- Single electron effects on transport (Coulomb blockade).




- Mechanical properties, surface effects and no dislocations



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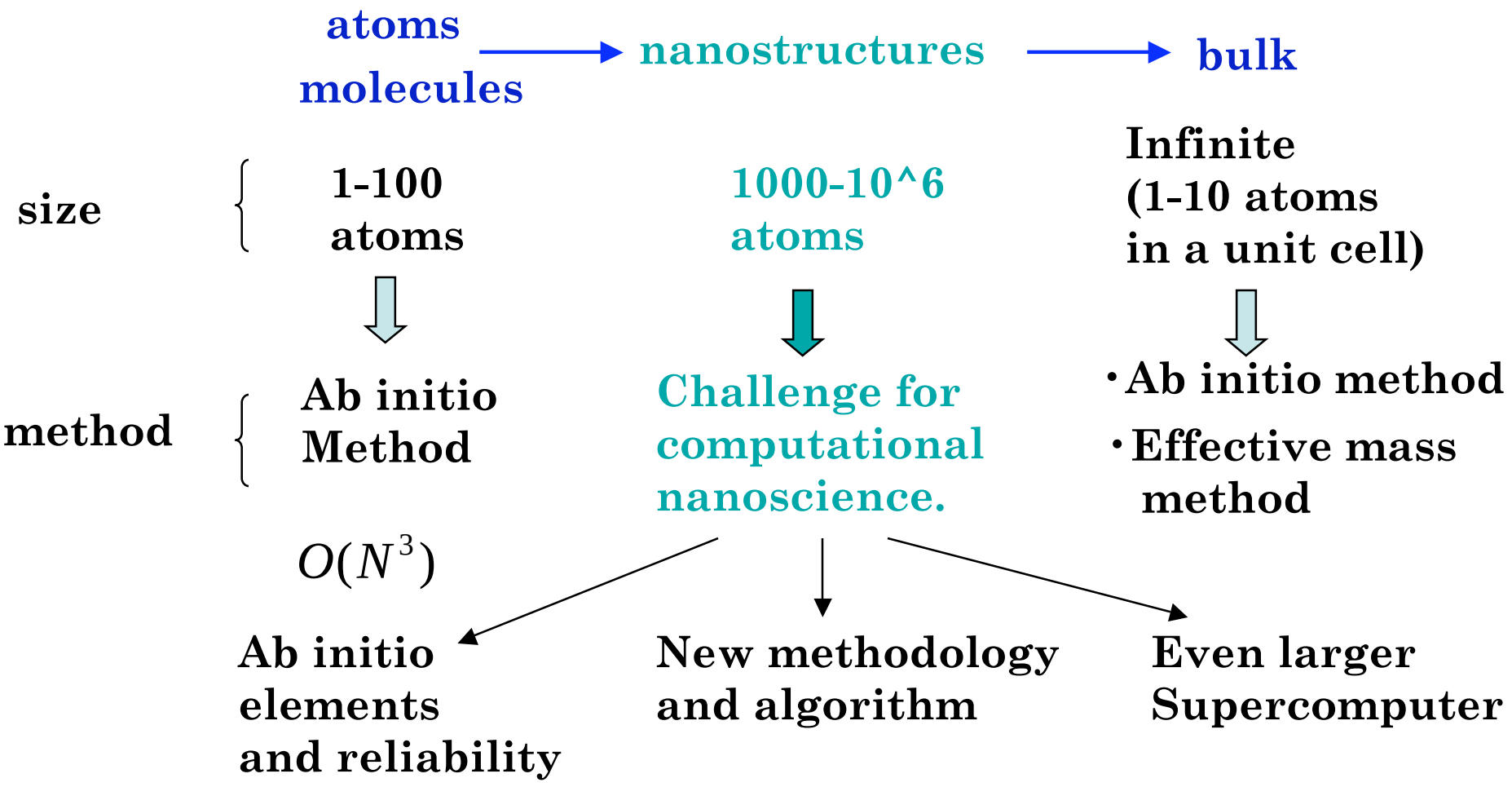
# Computational challenges (larger nanostructures)



# Ab initio Method: Density Functional Theory (Kohn 98 Nobel Prize)

Many Body Schrodinger Equation (exponential scaling )

$$\{-\sum_i \frac{1}{2}\nabla_i^2 + \sum_{i,j} \frac{1}{|r_i - r_j|} + \sum_{i,I} \frac{Z}{|r_i - R_I|}\}\Psi(r_1,..r_N) = E\Psi(r_1,..r_N)$$

**Kohn Sham Equation (65): The many body problem can be mapped onto a single particle problem with the same electron density and a different effective potential (cubic scaling cost).**

$$\{-\frac{1}{2}\nabla^2 + \int \frac{\rho(r')}{|r - r'|}dr' + \sum_I \frac{Z}{|r - R_I|} + V_{XC}\}\psi_i(r) = E_i\psi_i(r)$$

$$\rho(r) = \sum_i |\psi_i(r)|^2 = |\Psi(r_1,..r_N)|^2$$

Use Local Density Approximation (LDA) for $V_{XC}[\rho(r)]$ (good Si,C)

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# Plane-wave Pseudopotential Method in DFT

$$\{-\frac{1}{2}\nabla^2 + \int \frac{\rho(r')}{|r-r'|}dr' + \sum_I \frac{Z}{|r-R_I|} + V_{XC}(\rho(r))\}\psi_j(r) = E_j\psi_j(r)$$

**Solve Kohn-Sham Equations self-consistently for electron wavefunctions within the Local Density Appoximation**

1. **Plane-wave expansion for** $\psi_{j,k}(r) = \sum_g C_g^j(k) e^{i(g+k).r}$
2. **Replace "frozen" core by a pseudopotential**

**Different parts of the Hamiltonian calculated in different spaces (fourier and real) 3d FFT used**

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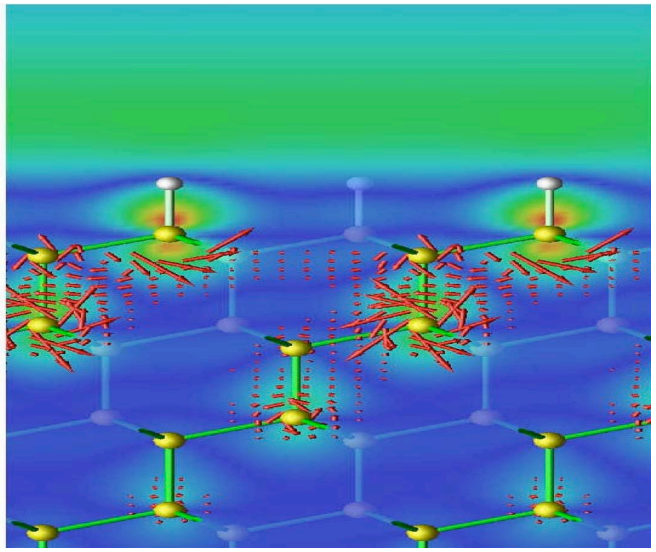
# CRD PARATEC (PARAllel Total Energy Code)



- PARATEC performs first-principles quantum mechanical total energy calculation using pseudopotentials & plane wave basis set
- Designed to run on large parallel machines IBM SP etc. but also runs on PCs

- PARATEC uses all-band CG approach to obtain wavefunctions of electrons
- Generally obtains high percentage of peak on different platforms
- Developed with Louie and Cohen's groups (UCB, LBNL), Raczkowski (Multiple 3d FFTs Peter Haynes and Michel Cote)

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# PARATEC: Performance

| Problem | P | NERSC (Power3) | | Jaquard (Opteron) | | Thunder (Itanium2) | | Phoenix (X1) | | NEC ES (SX6*) | | NEC SX8 | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | Gflops/P | %peak | Gflops/P | %peak | Gflops/P | %peak | Gflops/P | %peak | Gflops/P | %peak | Gflops/P | %peak |
| 488 Atom CdSe Quantum Dot | 128 | 0.93 | 62% | | | 2.8 | 51% | 3.2 | 25% | 5.1 | 64% | 7.5 | 64% |
| | 256 | 0.85 | 67% | 1.98 | 45% | 2.6 | 47% | 3.0 | 24% | 5.0 | 62% | 6.8 | 62% |
| | 512 | 0.73 | 49% | 0.95 | 21% | 2.4 | 44% | | | 4.4 | 55% | | |
| | 1024 | 0.60 | 40% | | | 1.8 | 32% | | | 3.6 | 46% | | |

- All architectures generally achieve high performance due to computational intensity of code (BLAS3, FFT)
- ES achieves highest overall performance to date: 5.5Tflop/s on 2048 procs
  - Main ES advantage for this code is fast interconnect
- SX8 achieves highest per-processor performance
- X1 shows lowest % of peak
  - Non-vectorizable code much more expensive on X1 (32:1)

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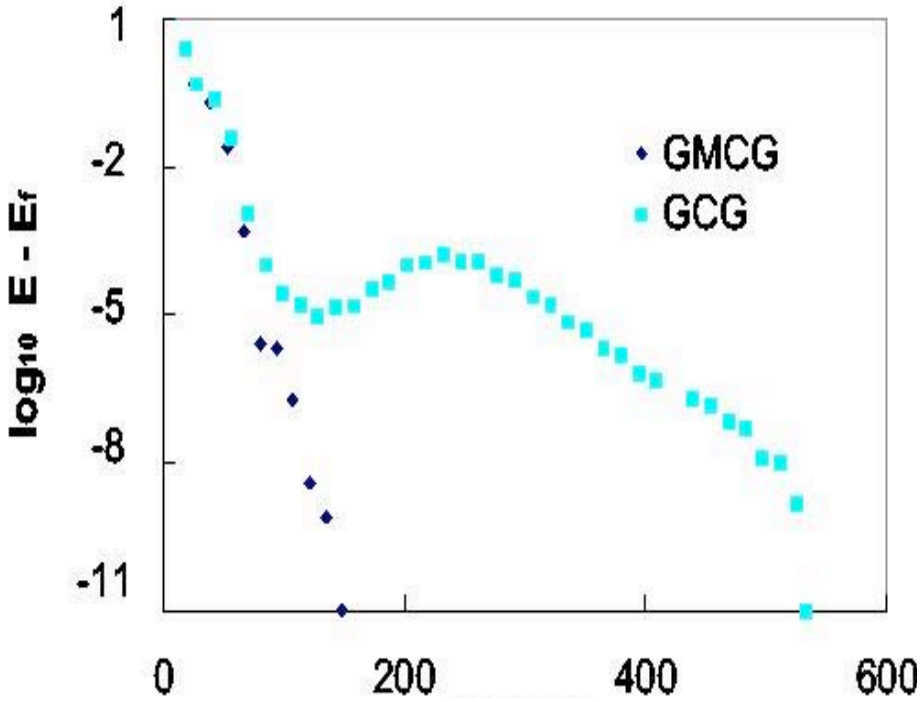
Developed with Louie and Cohen's groups (UCB, LBNL), also work with L. Oliker, J Carter

# Self-consistent all band method for metallic systems

Previous methods use self-consistent (SC) band by band, with Temperature smearing (eg. VASP code)

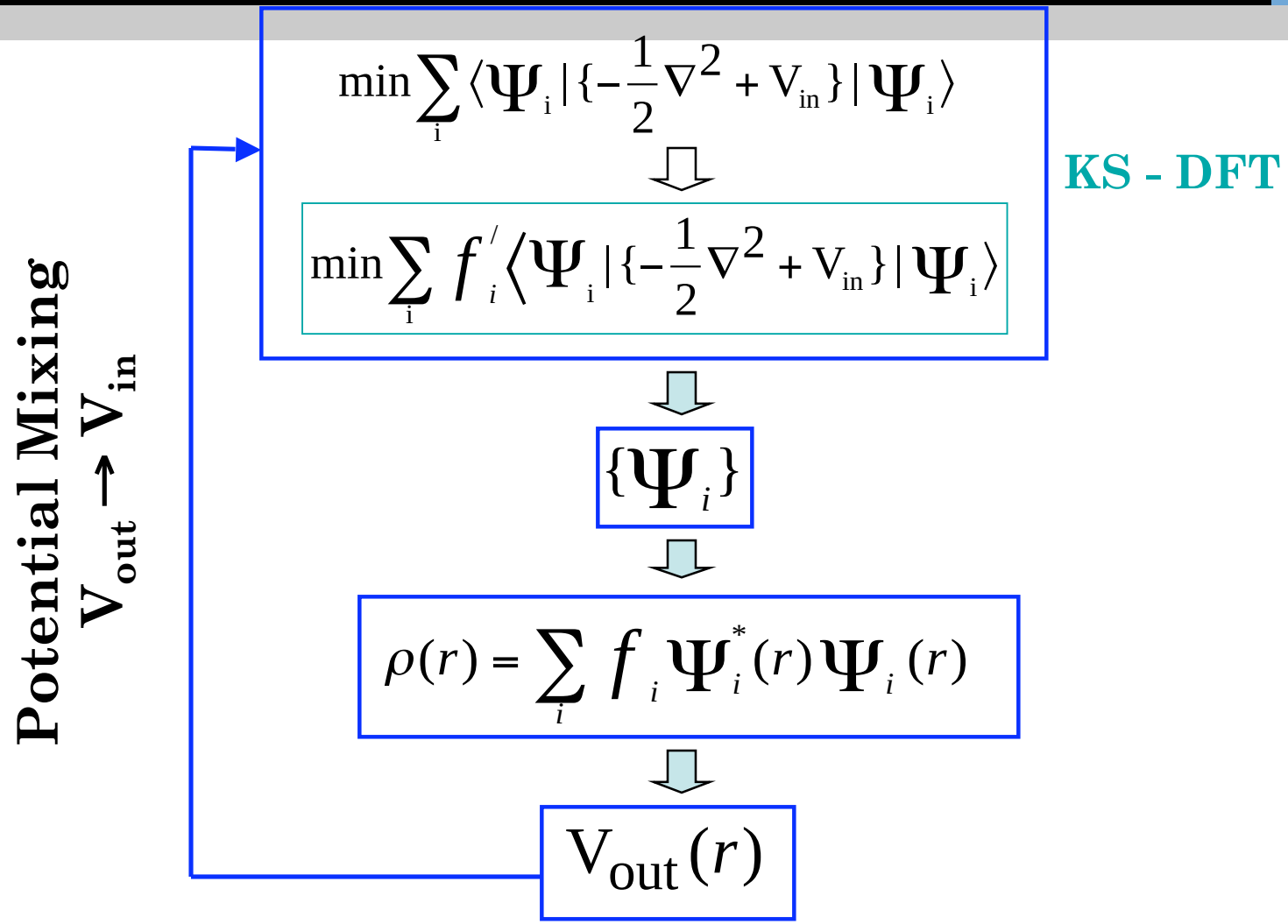
drawbacks – band-by-band slow on modern computers (cannot use fast BLAS3 matrix-matrix routines)

New Method uses occupancy in inner iterative loop with all band Grassman method (GMCG method)



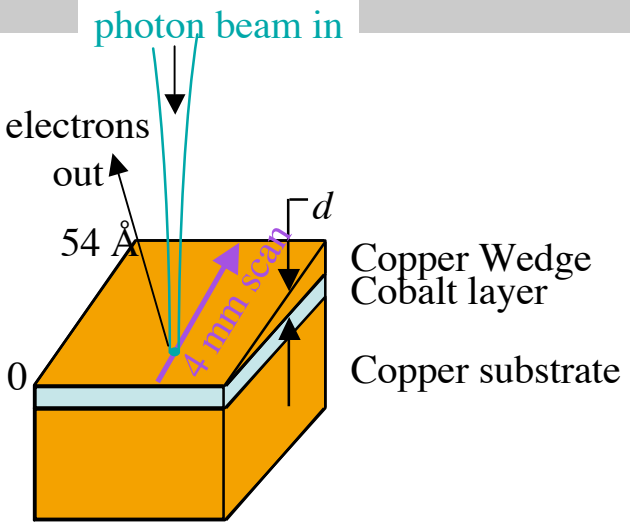

**Al (100) surface, 10 layers + vacuum**
**GMCG: new method with occupancy**

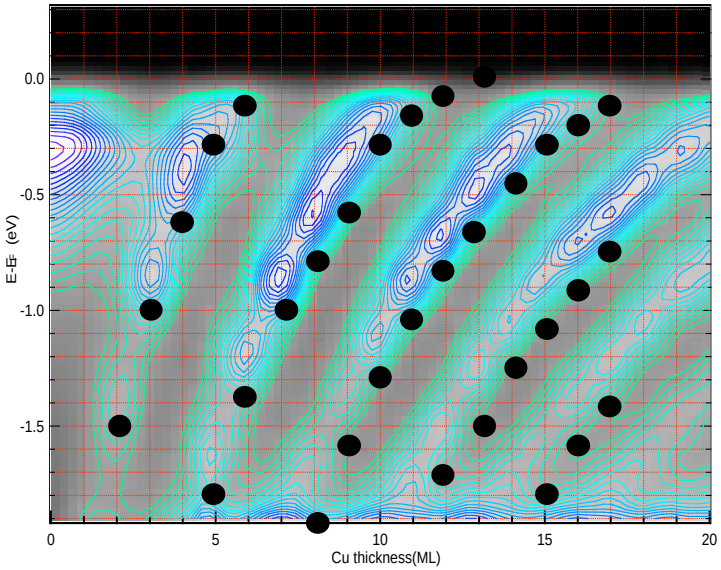
# Self-consistent all band method for metals

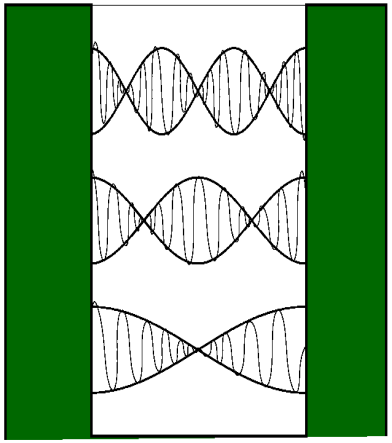


# The Quantization Condition of Quantum-well States in Cu/Co(100)

- **Theoretical investigation of Quantum Well states in Cu films using our codes (PARATEC, PEtot) to compare with experiments at the ALS (E. Rotenberg, Y.Z. Wu, Z.Q. Qiu)**
- **New computational methods for metallic systems used in the calculations.**
- **Lead to an understanding of surface effects on the Quantum Well States. Improves on simple Phase Accumulation Model used previously**



**QW states in Copper Wedge**

Difference between theory and experiment improved by taking surface effects into account

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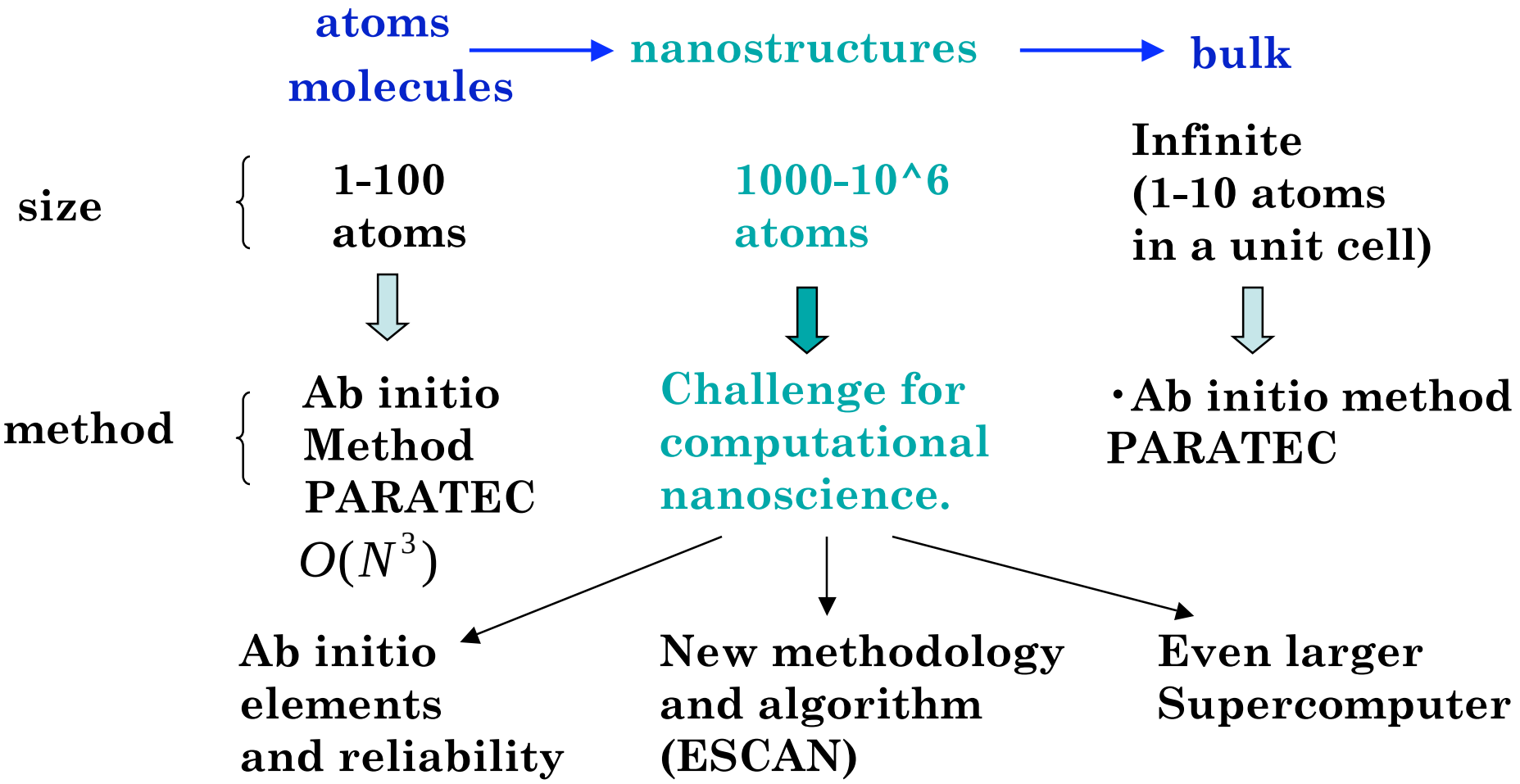
# Computational challenges (larger nanostructures)

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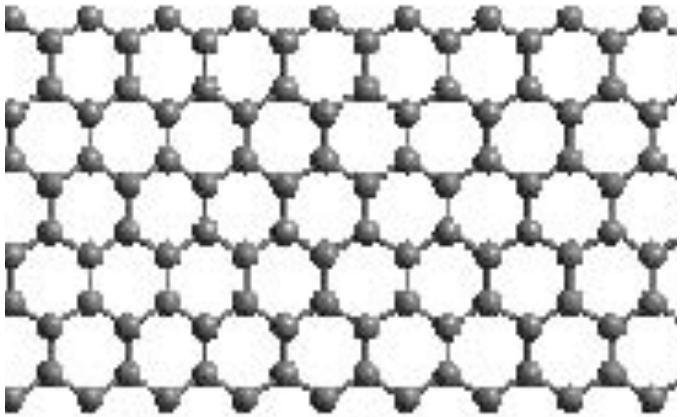
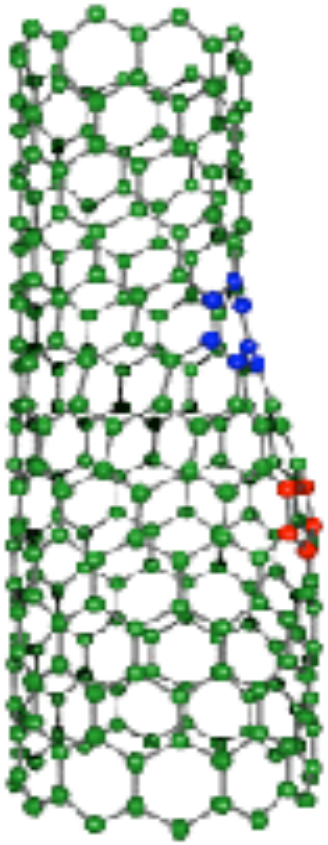
# Charge patching method for larger systems (Wan



**Selfconsistent LDA calculation of a single graphite sheet**



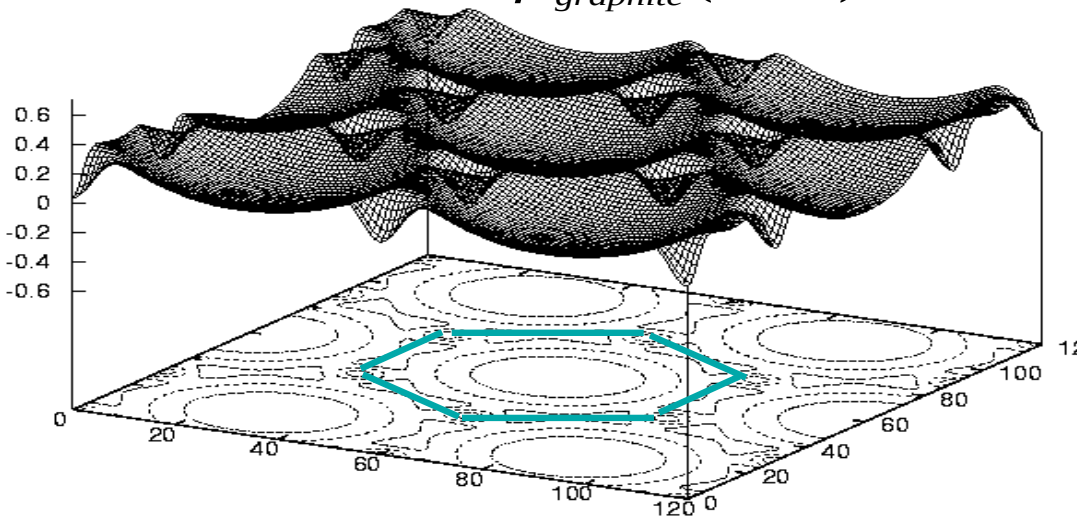
**Non-selfconsistent LDA quality potential for nanotube**

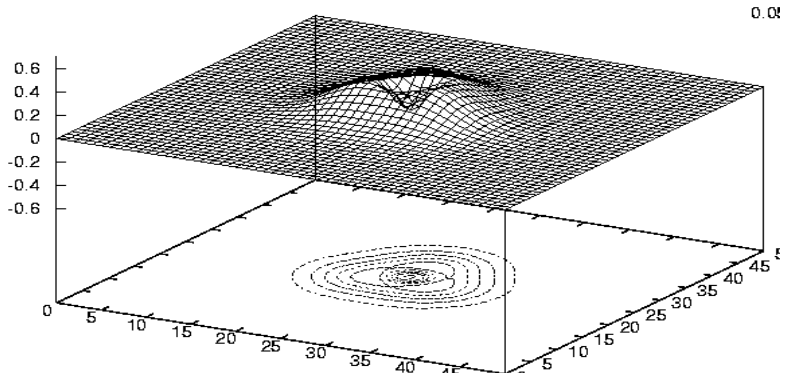


**Get information from small system ab initio calc., then generate the charge densities for large systems**

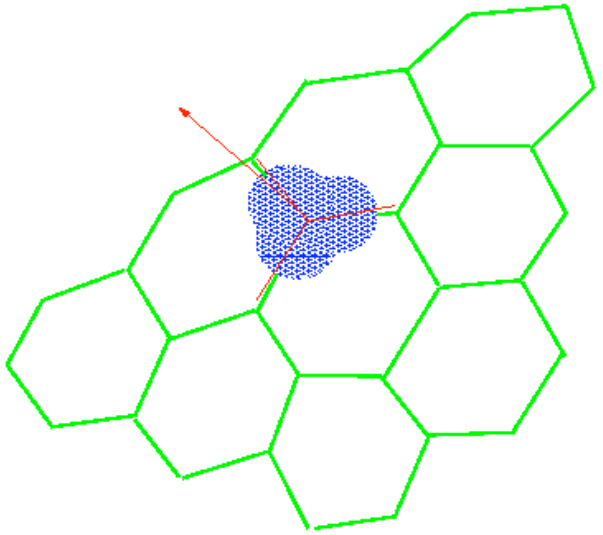
# Motif based charge patching method

$\rho_{graphite}(LDA)$ → $\rho_{motif}$




$$\rho^{patch}_{nanotube}(r) = \sum_{R} \rho^{aligned}_{motif}(r - R)$$



**Error: 1%, ~20 meV eigen energy error.**

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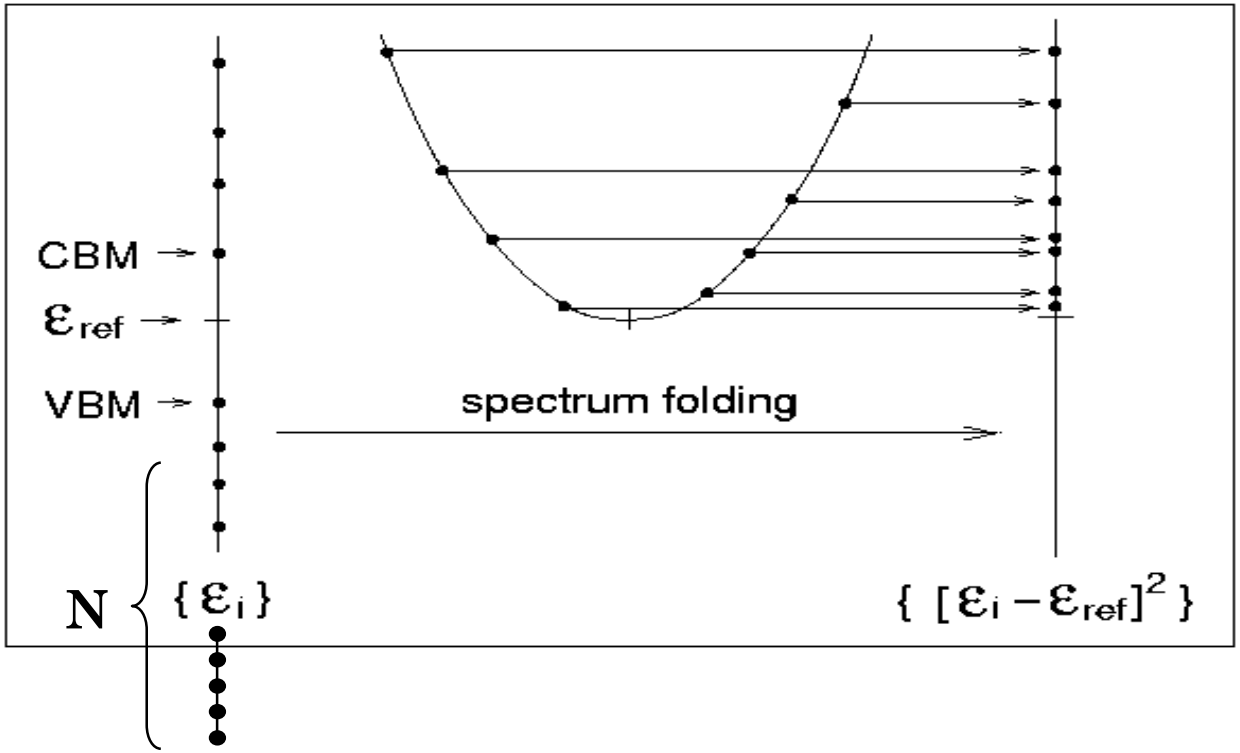
# + Folded Spectrum Method (ESCAN)

$$\{-\frac{1}{2}\nabla^2 + V(r)\}\psi_i(r) = E_i\psi_i(r)$$

$$H\psi_i = \varepsilon_i\psi_i \qquad (H-\varepsilon_{ref})^2\psi_i = (\varepsilon_i-\varepsilon_{ref})^2\psi_i$$



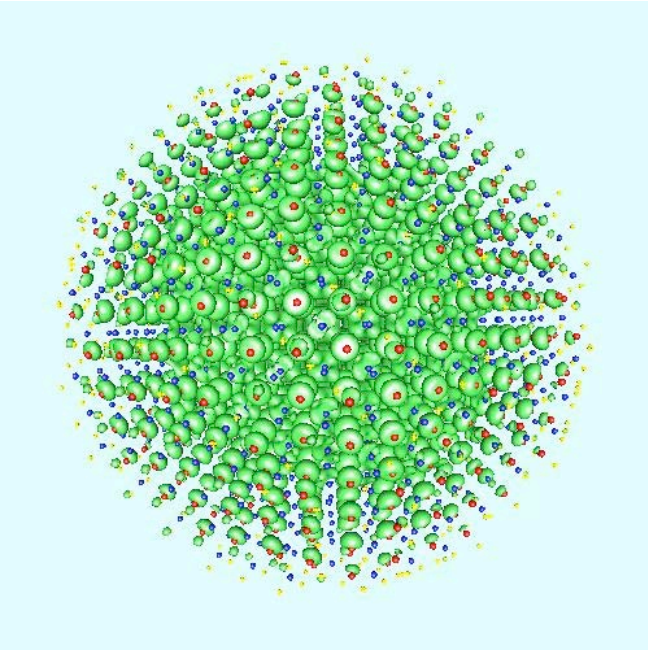
# Charge patching: free standing quantum dots

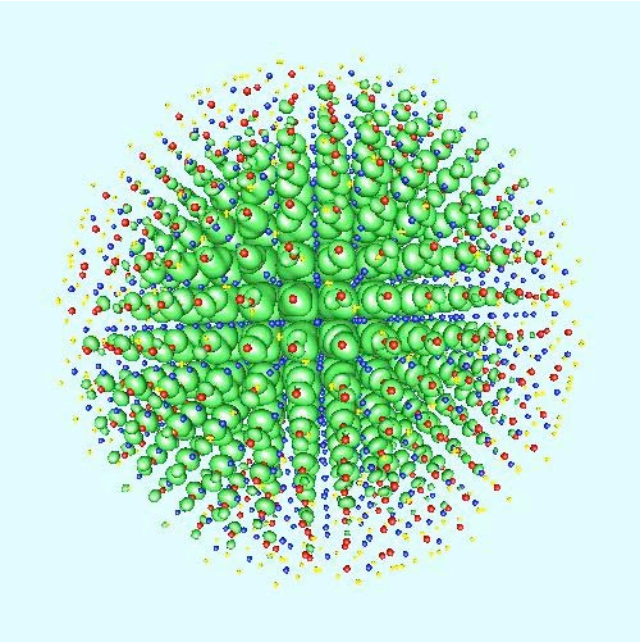


## $In_{675}P_{652}$ LDA quality calculations (eigen energy error ~ 20 meV)

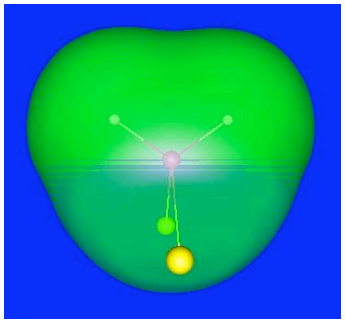
**64 processors (IBM SP3) for ~ 1 hour**

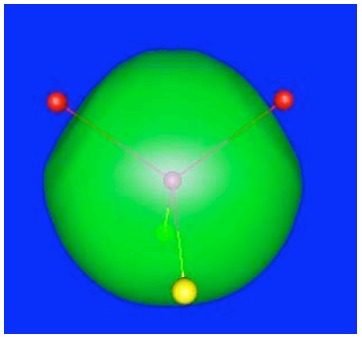
**CBM** **VBM**



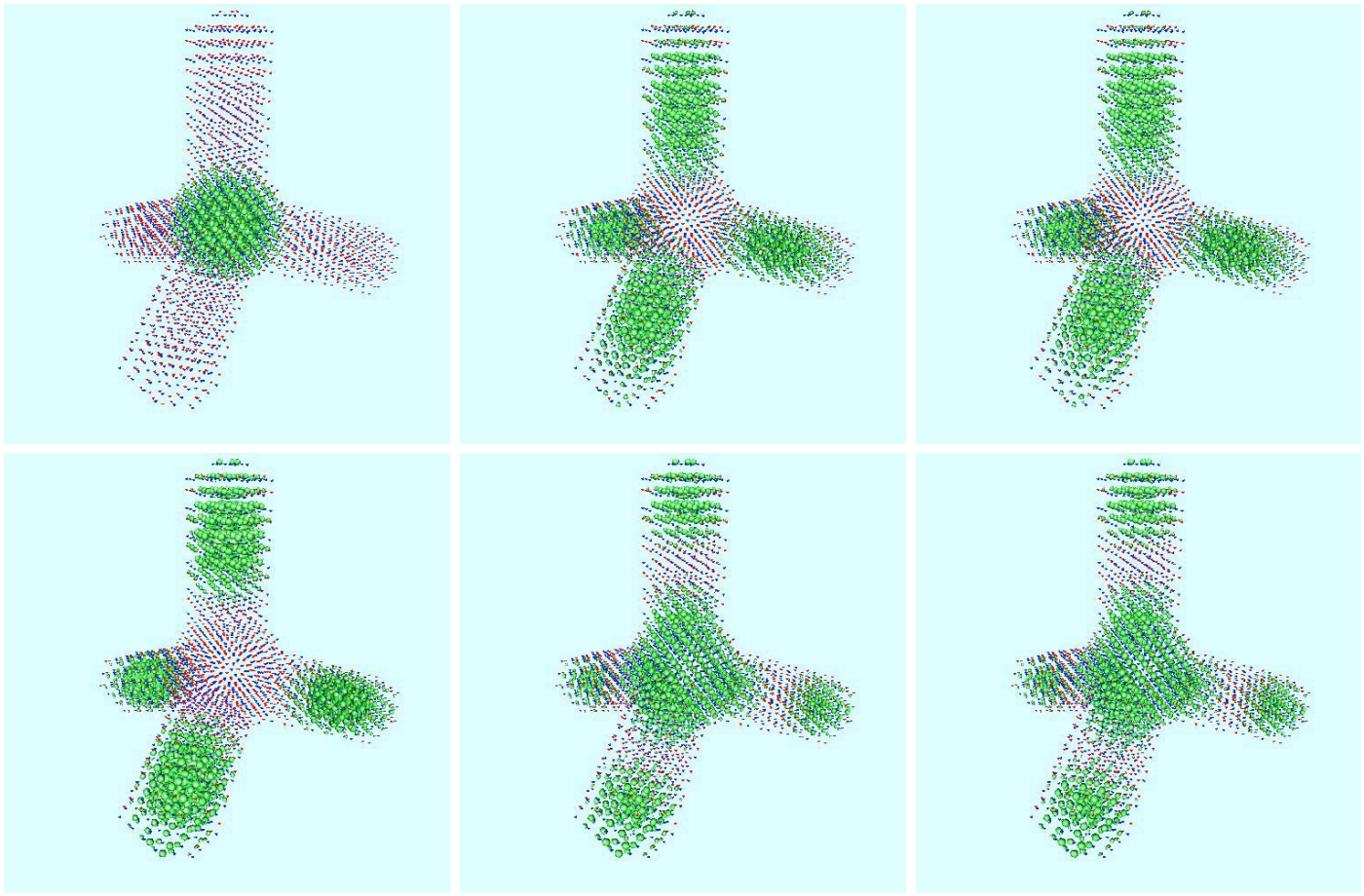


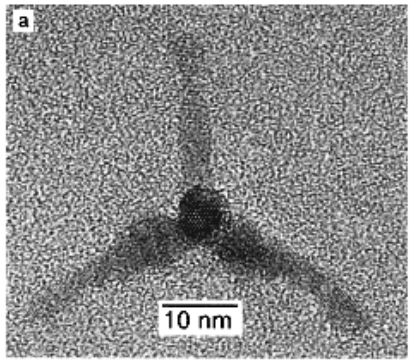
Total charge density motifs





# CdSe tetrapod electronic states

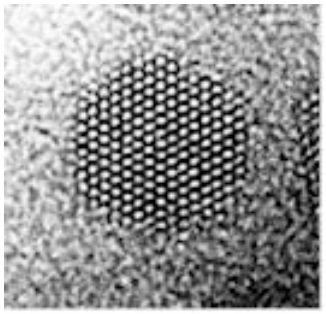





Alivisatos group UCB, LBNL

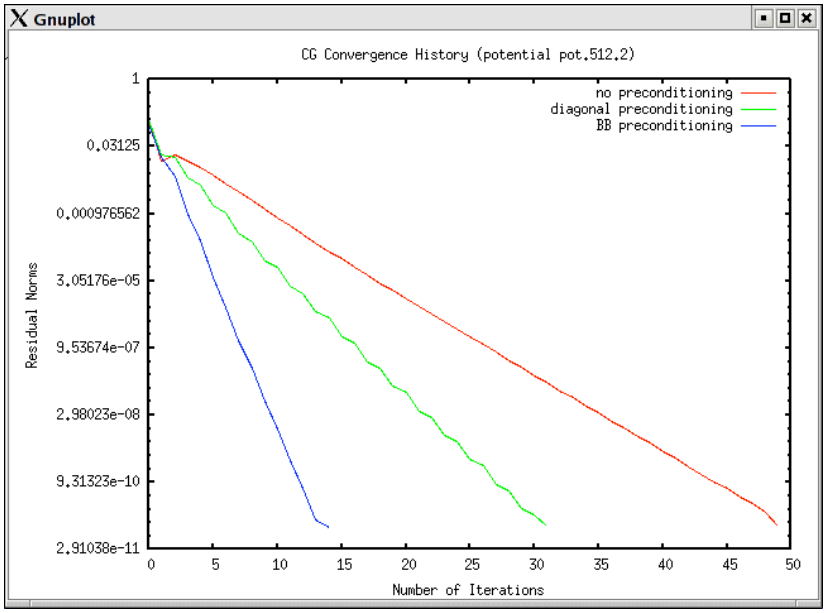
# CRD New Preconditioners and Eigensolvers

**Using preconditioner based on bulk states for nanosystems (away from the surface of the nanostructure system is bulk like)**



CdSe Quantum Dot

**Convergence of a 512 atom CdSe system with new preconditioner using the ESCAN code**

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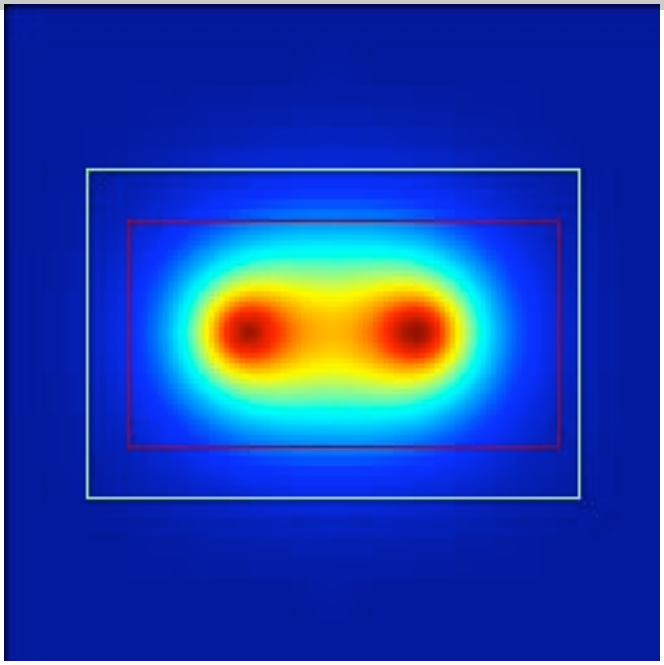
Work carried out by S. Tomov, J. Langou, C Voemel, L-W Wang

# AMR Density Functional Theory (J. Bell, C. Rendleman)

- Real-space methods for Kohn-Sham DFT
  - Electronic orbitals defined on AMR hierarchy
  - Higher-order, compact schemes
  - $O(\alpha MN^3)$ storage ($\alpha<1$ with AMR)
- Spectral methods restricted to fixed resolution grids
  - Pseudo-spectral, high order discretizations
  - $O(MN^3/2)$ storage
- AMR concentrates work where interesting physics occurs (atomic cores, bonds) and away from inter-atomic regions

# CRD AMR DFT Preliminary Results



| Effective Resolution | Time | Time-AMR | Refined Domain |
|---|---|---|---|
| $32^3$ | 20 | | |
| $64^3$ | 160 | 100 | 18.75 % |
| $128^3$ | 2237 | 290 | 7.3 % |

- Hydrogen molecule
  - 2 Levels of AMR
  - Approximately 10 iterations converge total energy.

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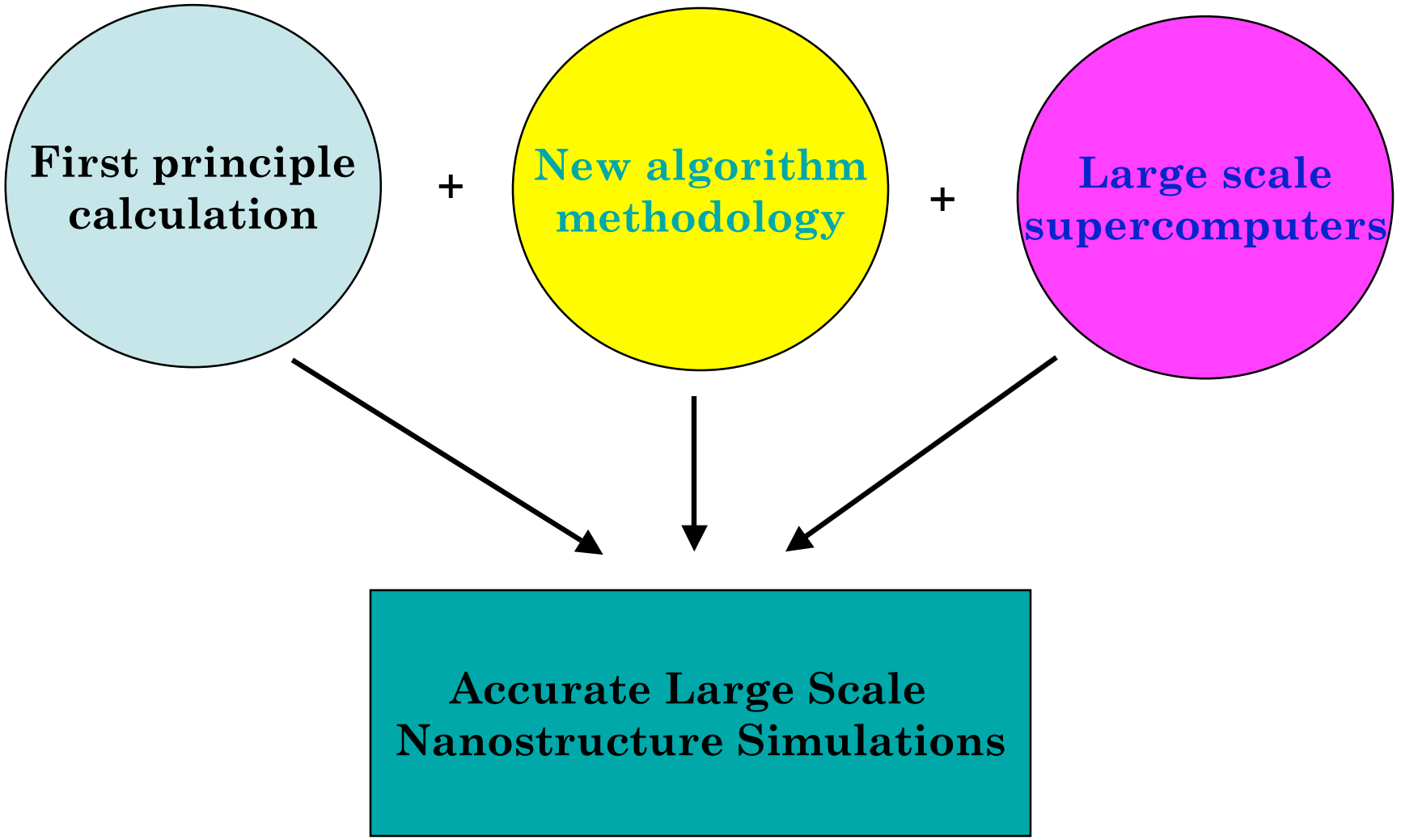
# Other projects



- **Screened Exchange method for accurate band gaps (improves on DFT)**
  **(L-W Wang, B. Lee, A. Canning)**
- **Generalized Pattern Search Methods for a Surface Structure Determination Problem**
  **(M. van Hove, J. Meza, Z. Zhao)**
- **A Constrained Optimization Algorithm for Total Energy Minimization in Electronic Structure Calculations**
  **(C. Yang, J Meza, L-W Wang)**


First principle calculation
+
New algorithm methodology
+
Large scale supercomputers
Accurate Large Scale Nanostructure Simulations